IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: Qiu *et al.* Appl. No. To be assigned (Divisional of U.S. Appl. No. 09/534,573; filed: March 27, 2000) Filed: January 18, 2002 For: **A Piezoelectric Element and Method of Manufacturing Same (as amended)**	Confirmation No.: Art Unit: To be assigned Examiner: To be assigned Atty. Docket: 1089.0350001/DKSC/TUM

Preliminary Amendment

Commissioner for Patents
Washington, D.C. 20231

Sir:

Please enter the following amendment into the application prior to the examination thereof. This Amendment is provided in the following format:

(A) A clean version of each replacement paragraph/section/claim along with clear instructions for entry; and

(B) Starting on a separate page, a marked-up version entitled: "Version with markings to show changes made."

Please amend the captioned application as follows:

Amendments

In the Title:

Please substitute the following Title of the Invention for the pending Title of the Invention: A Piezoelectric Element and Method of Manufacturing Same

In the Specification:

Please substitute the following paragraphs/sections for the pending paragraphs/sections.

Substitute the 1st full paragraph on page 7, with the following paragraph:

The present invention is a method for manufacturing a piezoelectric material having a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a" comprising a first step of producing an oxide containing an element "a' ", and a second step of producing a piezoelectric material by subjecting the oxide containing the element "a' " produced in the first step to a hydrothermal process using an aqueous solution containing the element "a", wherein the amount of element "a" contained in the piezoelectric material produced in the second step is increased due to its substitution for element "a' " contained in the oxide produced in the first step.

Substitute the 1st full paragraph on page 8, with the following paragraph:

The oxide produced in the first step may be a piezoelectric material expressed by the formula ABO_3 in which the symbol A represents at least an element "a'", and having a perovskite crystal structure. This allows the hydrothermal process of the second step to be performed for the piezoelectric material that has already been crystallized in a perovskite crystal structure, and allows the element "a' " at the A site of this piezoelectric material to be substituted with the element "a".

Substitute the third paragraph on page 11 that continues to the top of page 12, with the following paragraph:

In another method of the present invention for manufacturing a piezoelectric material, the element "a" is barium, the element "a' " is lead, and the element expressed by the symbol B is titanium. Furthermore, the oxide produced in the first step is preferably lead titanate ($PbTiO_3$) composed of acicular crystals. It is also preferable if the first step produces the oxide by Metal-Organic Deposition (hereinafter "MOD"). This makes it possible to manufacture a piezoelectric material that is $BaTiO_3$ composed of acicular crystals with a large grain size, and more accurately, barium-lead titanate expressed by the chemical formula $(Ba_xPb_{1-x})TiO_3$, where x in the formula is within the range of $0 < x < 0.05$, which was difficult to manufacture with a conventional method.

Substitute the first full paragraph of page 12, with the following paragraph:

The piezoelectric material of the present invention is expressed by the chemical formula (Ba, Pb)TiO_3, wherein the piezoelectric material is composed of acicular crystals, at a specific spacing there are dislocation layers in which lattice defects are present, and the spacing between adjacent dislocation layers is at least 10 nm. This makes it possible to provide barium-lead titanate with excellent voltage resistance and good piezoelectric characteristics. In particular, it is possible to provide barium-lead titanate expressed by the chemical formula $(Ba_xPb_{1-x})TiO_3$, where x in this formula is within the range of $0 < x < 0.05$.

Substitute the first full paragraph of page 24, with the following paragraph:

A piezoelectric film precursor with a thickness of 0.4 μm and comprising four laminated layers is obtained in the above process. In addition to the sol-gel method discussed above, the step of forming the piezoelectric film precursor can also be accomplished by RF sputtering, ion beam sputtering, MOD (Metal-Organic Deposition), electron beam vapor deposition, or another such method.

Substitute the first full paragraph of page 37, with the following paragraph:

A piezoelectric actuator, which is a piezoelectric element, equipped with a piezoelectric material expressed by the compositional formula $Pb_xBa_{(1-x)}(Zr_{56}Ti_{44})O_3$ was manufactured according to the above third manufacturing method. Figure 13 is a graph of the EDX spectrum piezoelectric precursor film as the composition before hydrothermal processing. As a comparative example of a conventional method, Figure 10 is a graph of the EDX spectrum of the final product when the hydrothermal processing was performed with an alkali aqueous solution of just $Ba(OH)_2$ (molar ratio of barium and lead = 1:0). As Example 1 of the present invention, Figure 11 is a graph of the EDX spectrum of the final product when the hydrothermal processing was performed with an alkali aqueous solution in which the molar ratio of barium and lead was adjusted to 6:4. As Example 2 of the present invention, Figure 12 is a graph of the EDX spectrum of the final product when the hydrothermal processing was performed with an alkali aqueous solution in which the molar ratio of barium and lead was adjusted to 4:6.

Substitute the third paragraph on page 40 that continues to the top of page 41, with the following paragraph:

It is generally preferable for the crystal grains to be large and acicular in order to enhance the piezoelectric characteristics of a piezoelectric material. However, it is difficult to produce large, acicular crystal grains from barium titanate ($BaTiO_3$), and the crystal grains turn out small when manufactured by MOD or a sol-gel method, for example. In contrast, it is relatively easy to produce large-diameter, acicular crystal

grains by MOD from lead titanate ($PbTiO_3$). In view of this, if a crystalline material of lead titanate is produced by MOD, and is then subjected to a hydrothermal processing with a barium hydroxide aqueous solution, at least 95% of the A sites can be substituted with barium. Specifically, it is possible with this manufacturing method to produce lead-barium titanate expressed by the chemical formula $(Ba_xPb_{1-x})TiO_3$, where x in this formula is within the range of $0 < x < 0.05$.

In the Claims:

Please cancel claims 1-18, and 21-26 without prejudice to or disclaimer of the subject matter therein.

Please substitute the following claims 19, 20, and 27-29 for the pending claims 19, 20, and 27-29.

19. (Amended) A method for manufacturing a piezoelectric element having a piezoelectric material with a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a", comprising the steps of:

a) forming a lower electrode;

b) forming over the lower electrode a film of the piezoelectric material having a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a" by,

a first step of producing an oxide in an amorphous state containing an element "a' " and subjecting the oxide to a hydrothermal process using an aqueous solution containing the element "a' " thereby crystallizing the oxide, wherein the oxide produced in the first step is a piezoelectric material having a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a' "; and

a second step of producing a piezoelectric material by subjecting the oxide produced in the first step to a hydrothermal process using an aqueous solution containing the element "a", so as to increase the amount of the element "a" contained in the piezoelectric material due to its substitution for element "a' " contained in the oxide produced in the first step; and

c) forming an upper electrode over the piezoelectric material formed in step b.

20. (Amended) A method of forming an ink-jet recording head, comprising the steps of:

forming a diaphragm film over a substrate;

manufacturing a piezoelectric element over the diaphragm film by the method for manufacturing a piezoelectric element according to claim 19; and

working the substrate and forming a pressurization chamber at a site capable of transmitting displacement of the diaphragm film produced by driving of the piezoelectric element.

27. (Amended) A piezoelectric element comprising:

a piezoelectric material expressed by the chemical formula $(Ba,Pb)TiO_3$, wherein the piezoelectric material is composed of acicular crystals having dislocation layers in which lattice defects are present and wherein the spacing between adjacent dislocation layers is at least 10 nm; and

electrodes with which voltage can be applied to said piezoelectric material.

28. An ink jet recording head, wherein the piezoelectric element according to claim 27 is provided as a piezoelectric actuator.

29. A printer, equipped with the ink jet recording head according to claim 28 as printing means.

Please add the following new claims 30-34.

30. (new) A method for manufacturing a piezoelectric element having a piezoelectric material with a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a", comprising the steps of:

a) forming a lower electrode;

b) forming over the lower electrode a film of the piezoelectric material having a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a" by,

a first step of producing an oxide in an amorphous state containing an element "a' "; and

a second step of producing a piezoelectric material by crystallizing the oxide produced in the first step in a hydrothermal process using an aqueous solution containing the element "a", so as to increase the amount of the element "a" contained in the piezoelectric material due to its substitution for element "a' " contained in the oxide produced in the first step; and

c) forming an upper electrode over the piezoelectric material formed in step b.

31. (new) The method for manufacturing a piezoelectric element according to claim 30, wherein the hydrothermal process performed in step b is conducted using an aqueous solution containing both the element "a" and the element "a' ", and wherein the ratio in which the element "a" and the element "a' " are present in the aqueous solution is between 2:8 and 4:6.

32. (New) The method for manufacturing a piezoelectric element according to claim 31, wherein the aqueous solution containing the element "a' " is an alkali aqueous solution of a compound expressed by the formula $a'(OH)_n$ (n = 1, 2, or 3).

33. (New) The method for manufacturing a piezoelectric element according to claim 30, wherein the oxide in an amorphous state produced in step b is produced by pyrolyzing a sol containing an organometallic.

34. (New) The piezoelectric element according to claim 27, wherein said piezoelectric material is expressed by the chemical formula $(Ba_xPb_{1-x})TiO_3$, and x in this formula is within the range of $0 < x < 0.05$.

In the Drawings:

Please substitute Figure 3 which accompanies the ***Request to Approve Proposed Drawing Corrections*** submitted herewith for the pending Figure 3.

Remarks

Consideration of this Application is respectfully requested.

Upon entry of the foregoing amendment, claims 19, 20, 27-29 and 30-34 are pending in the application, with 19, 27 and 30 being the independent claims. Claims 19, 20, and 27 have been amended. Further, a clean version of claims 28 and 29 is included in compliance with 37 C.F.R. §1.121(c)(3). Claims 30-34 have been added. The amendment of the specification and revision to Figure 3 address rejections made by the Examiner during the prosecution of the parent application, *i.e.*, U.S. Appl. No. 09/534,573, filed March 25, 2000 (allowed). These changes are believed to introduce no new matter, and their entry is respectfully requested.

Prompt and favorable consideration of this Preliminary Amendment is respectfully requested.

Respectfully submitted,

STERNE, KESSLER, GOLDSTEIN & FOX P.L.L.C.

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Version with markings to show changes made

Claims 19, 20 and 27 have been amended as follows.

19. (Amended) A method for manufacturing a piezoelectric element having a piezoelectric material with a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a", comprising the steps of:

a) forming a lower electrode;

b) forming over the lower electrode a film of the [a] piezoelectric material [exhibiting piezoelectricity by the method of manufacturing a piezoelectric material according to Claims 1-7] having a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a" by,

a first step of producing an oxide in an amorphous state containing an element "a' " and subjecting the oxide to a hydrothermal process using an aqueous solution containing the element "a' " thereby crystallizing the oxide, wherein the oxide produced in the first step is a piezoelectric material having a perovskite crystal structure expressed by the formula ABO_3 in which the symbol A represents at least an element "a'"; and

a second step of producing a piezoelectric material by subjecting the oxide produced in the first step to a hydrothermal process using an aqueous solution containing the element "a", so as to increase the amount of the element "a" contained in the piezoelectric material due to its substitution for element "a' " contained in the oxide produced in the first step; and

c) forming an upper electrode over the piezoelectric material formed in step b.

20. (Amended) A method of forming an ink-jet recording head, comprising the steps of:

forming a diaphragm film over a substrate;

manufacturing a piezoelectric element over the diaphragm film by the method for manufacturing a piezoelectric element according to [any of Claims 1 to 7] claim 19; and

working the substrate to form [and forming] a pressurization chamber at a site capable of transmitting displacement of the diaphragm film produced by driving of the piezoelectric element.

27. (Amended) A piezoelectric element comprising:

a [the] piezoelectric material [according to Claim 25 or 26] expressed by the chemical formula $(Ba,Pb)TiO_3$, wherein the piezoelectric material is composed of acicular crystals having dislocation layers in which lattice defects are present and wherein the spacing between adjacent dislocation layers is at least 10 nm; and

electrodes with which voltage can be applied to said [this] piezoelectric material.

Claims 30-34 were added to the application.

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Qui *et al.*	Art Unit: To Be Assigned
Appl. No. To Be Assigned (Divisional of U.S. Appl. No. 09/534,573; Filed: March 27, 2000)	Examiner: To Be Assigned
Filed: January 18, 2002	Atty. Docket: 1089.0350001/DKSC/TUM
For: **A Piezoelectric Element and Method of Manufacturing Same (as amended)**	

Request to Approve Proposed Drawing Correction

Commissioner for Patents
Washington, D.C. 20231

Sir:

Attached is a copy of one (1) sheet of drawings, containing proposed corrections to Figure 3, shown in red. The proposed changes add no new matter to this application.

Applicants request that the Examiner approve the proposed corrections.

Respectfully submitted,

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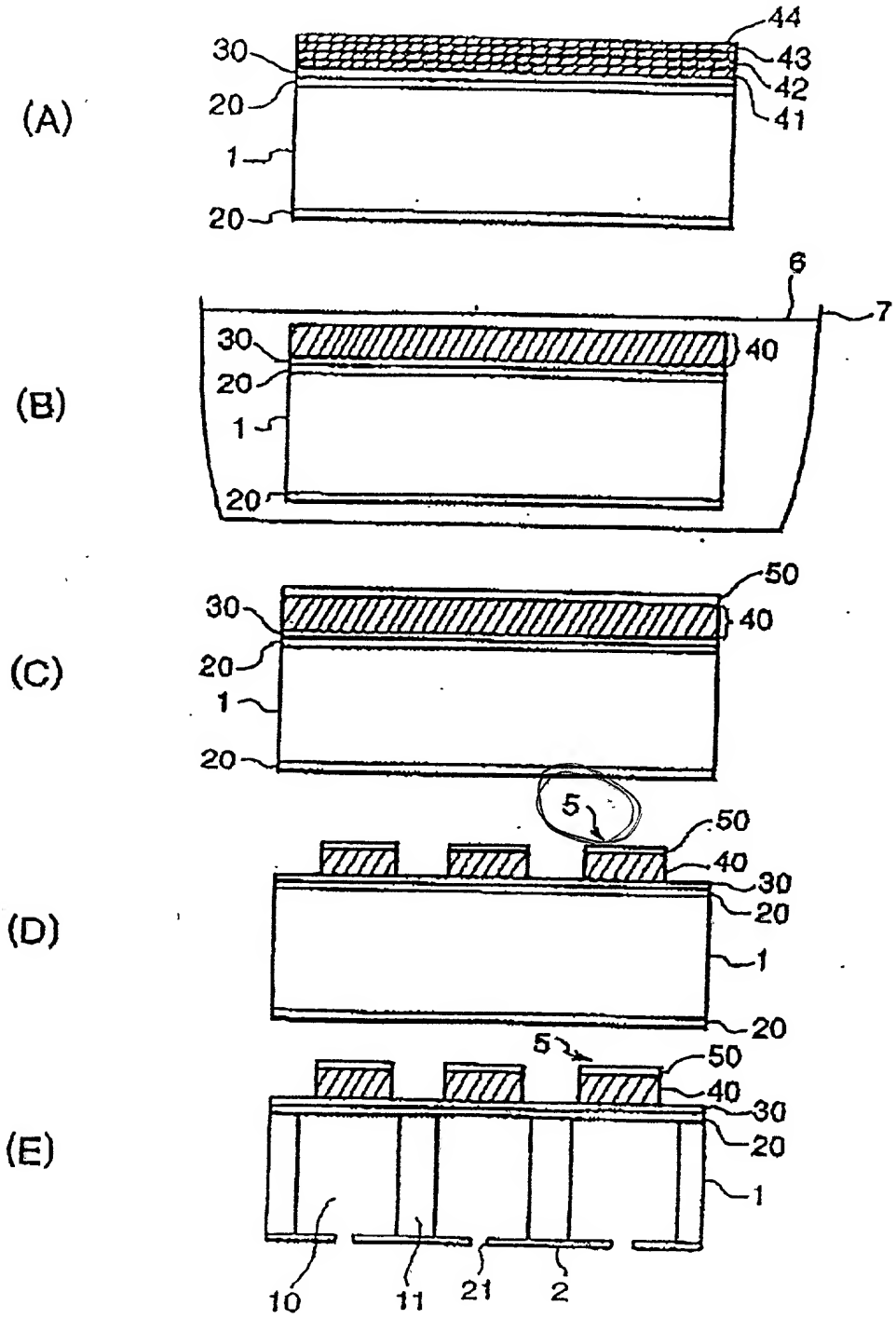
Fig. 3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